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Davidite and Other Early Events in Australia's Uranium Story

DAVID BRANAGAN

Abstract: The uranium-bearing mineral *davidite* was named for T.W. Edgeworth David. The controversy about its validity as a true mineral lasted some years. Significant studies of radioactivity and age determinations were carried out at Sydney University during the years 1904 to 1930.

Keywords: Edgeworth David, davidite, age determinations, Sydney University

INTRODUCTION

With the present interest in the use of uranium as a source of energy, it is an appropriate time to present a few snippets of early Australian research on that subject. The first scientific studies in Australia on radioactivity were carried out more than one hundred years ago. Although not himself an experimenter in this field, T.W. Edgeworth David (1858–1934), Professor of Geology at the University of Sydney between 1891 and 1924, through his encouragement and support of various students, played a significant part in the development of this research. Archibald Liversidge (1847–1927), Professor of Chemistry until 1907, also played an important role.

EDGEWORTH DAVID AND URANIUM

David and uranium are inextricably linked through the mineral davidite, named by Douglas Mawson (1882–1958) for his mentor, in 1906 (Mawson 1906). Mawson recognised the new material when he was involved in the examination of samples collected from the locality he named Radium Hill, in South Australia. The presence of carnotite (a yellow potassium uranyl vanadate), first attracted attention, as a probable decomposition product of another uranium compound. The primary substance in the orebody was black and sub-metallic. At first this main black material was suggested to be a single phase, probably a variety of ilmenite, but Mawson thought there were in fact five quite different substances, and he suspected there might be a new mineral present. Alderman (1967) makes the point that Mawson had not only a deep interest in minerals, but also an encyclopaedic knowledge. 'He displayed the hallmark of the great mineralogist–that uncanny ability to recognise almost instantly whether a mineral is unusual or "new" '.

Mawson described the 'new' substance as cuboid crystals of a black mineral with specific gravity about 4, having a brilliant lustre and glassy fracture, containing over 50% of TiO_2, a large quantity of iron and a notable amount of rare earths, uranium, vanadium and chromium. He continued 'the bright black mineral is an entirely new type, though details are not yet available for complete description. We propose to name it davidite, after Professor T.W.E. David, of Sydney University, whose personal ability, wise counsel, and enthusiasm have done so much to further the interests of the science and economic application of geology in Australasia.'

The mineral was supposed to have been chemically analysed by E.H. Rennie (1852–1927), a pupil and friend of Liversidge, (Barker & Stranks 1988), and his colleague W.T. Cooke (1877–1957) at Adelaide University, just arrived back from post-graduate studies in England. However, if their 'preliminary note' (1906) is right they did not test the correct material! They examined the material which Mawson wrote was associated with his 'new' mineral, but which was somewhat heavier and less brilliant in lustre. They pointed out that analysis was difficult, and to date incomplete, but that 'in addition to titanic and ferric oxides, which are the chief constituents, there are present uranium, vanadium, cerium, and almost certainly thorium and other rare earths, traces of lime, and we believe, also chromium and traces of manganese. The quantities of vanadium and chromium, however, if present, are very small, and in the presence of uranium difficult to detect with certainty' (Rennie & Cooke 1906).

The new mineral received its first international mention in the *Mineralogical Magazine* of September 1907, where L.J. Spencer (1870–1959) listed it among newly described minerals from around the world, commenting it was 'an incompletely described mineral, possibly identical with ilmenite'. Spencer mentioned that it was named for David. However the British establishment was not too satisfied with the validity of davidite as a mineral. Thus a sample was called for and sent by Mawson to the Imperial Institute in London, to be examined by two well-accredited members of the Scientific and Technical Department of the Institute, T. Crook and G.S. Blake (Crook & Blake 1910). They were sceptical, and while accepting that the complex substance might be new, after testing they wrote 'the existence of new minerals should only be inferred as a last resource to meet difficulties which are otherwise unmanageable. In the present instance, the evidence to be handled undoubtedly presents serious difficulties, but it seems less objectionable to cover this evidence by an appeal to known minerals than by an invention of new ones, ... pending the publication of proof to the contrary, one may reasonably continue to regard "davidite" as a mineral complex ... '

At the end of the paper Crook added a footnote that Mawson, back from the Antarctic (see below), had displayed specimens of 'davidite' at a meeting of the Geological Society of London on 9 February 1910, saying it was a 'homogeneous mineral', and claiming that Crook and Blake had not seen it. Mawson gave specimens to Crook several days later, saying that 'full details of chemical analyses by Drs. Rennie and Cooke will be published shortly'. However Crook stuck to his guns, saying that he couldn't see any difference from what they had tested, writing 'the mere uniformity of appearance and continuity of a fracture-surface (Mawson believed it was a crystal face) is not sufficient to prove that the material is homogeneous; and the fragments of "davidite" which we examined show unmistakeable signs of heterogeneity' (Crook & Blake 1910).

The years from 1906 to 1910 had not been conducive to Mawson expanding on his new mineral, as David gained him a position on Shackleton's Antarctic expedition and the two of them went off to seek the South Magnetic Pole, which incidentally kept the both of them interested, at times, in mineralogy (Branagan, 2005).

It was thus some time before Mawson came back to defend his mineral. By 1911 the orebody at Radium Hill had been opened up to some degree and Mawson felt 'further reference to the association and identity of davidite is due. The davidite in its pure form is but rarely met within the lode' (Mawson 1916). He had written a reply to Crook and Blake some time in 1911 with an attendant chemical analysis, but the paper was mislaid during the period preparing for his own Antarctic expedition departure in December 1911. The paper finally saw the light in August 1916 (Mawson 1916), not long after Mawson had gone to England again to work for the Allies' war effort.

In this 1916 paper Mawson claimed that the British researchers had also, like the original 'analysis' by Rennie and Cooke (1906) tested the wrong material! Mawson's paper was accompanied by an analysis by Dr. Cooke (of Adelaide), following up on the earlier analytical work by Rennie and himself (Rennie must have been too busy to continue with the analytical work, as had been proposed, so it was left to Cooke). Cooke was sure of himself, and wrote, 'of the ferriferous and titaniferous radio-active constituents of the lode, the one referred to as davidite is the most interesting, as it is homogeneous and is a distinct species.' He gave the following analysis: TiO_2 54.3, FeO 16.0, Fe_2O_3 13.0, rare earths 8.3, V_2O_5, Cr_2O_3, and U_2O_8 4.6, MgO 0.6, CaO 1.5, PbO 1.1, CuO trace, H_2O 1.5, Total 100.9. Mawson believed himself totally vindicated.

Mawson's 1916 paper and the attendant analysis by Cooke were probably only in press when Charles Anderson (1876–1944) of the Australian Museum, Sydney, completed his extensive, important bibliography of Australian mineralogy, so it is not surprising that Anderson (1916), relying apparently on the sceptical 1910 paper by Crook and Blake (1910), surrounded 'davidite' with inverted commas.

It was more than thirty years before Cooke's analysis was supported by other tests. There was revived interest in uranium in South Australia just as World Waw Two was ending (Mawson 1944, Mudd 2005). Of more significance for the present subject was the discovery in Mozambique in 1950 of a mineral apparently akin to, or closely related to, davidite (Bannister & Horne 1950). These researchers devoted a considerable time studying the mineral structure and testing comparable material from Radium Hill, indicating a close similarity between the samples from two such widely separated localities. The Mozambique region was shortly after looked at in considerable detail by Davidson & Bennett (1950). Bannister & Horne (1950) also pointed out an obscure reference (Golubkova 1930) indicating that a davidite-like substance had also been discovered in Russia somewhat earlier. Other sources of davidite were later discovered around the world.

Economic interest in radioactivity had remained largely centred on radium until the 1930's. In 1951 demand for uranium increased and attention turned once again to Radium Hill. Renewed exploration of the mine indicated a substantial deposit and it was realised that the significant uranium ore mineral was davidite (Nininger 1954). Almost one million tonnes of davidite was mined over the next few years at Radium Hill with an average ore grade of 1.2 kg/U_3O_8 per ton. The South Australian Geological Survey began an extensive study of uranium occurrences (Dickinson et al. 1954). Interest in uranium in NSW was also considerable between 1954 and 1961 (Mulholland & Rayner 1953, Rayner 1955, 1957).

During the South Australian Survey work detailed mineralogical examinations were carried out by the petrologist Alick Whittle (1920–1987), who made a considerable study of davidite (Whittle 1954). The work indicated the complexity of its structure and there is a hint that he suspected it was not a true mineral but a metamict phase. Rayner (1957), relying to some extent on Whittle, expanded on this matter, indicating that davidite appeared to be 'devoid of internal regular atomic arrangement', and there were 'no X-ray diffraction patterns of crystalline material'. In 1959, Whittle came out strongly that davidite was not a true mineral but a complex mixture (Whittle 1959). However the cudgel for its validity was taken up by several North American mineralogists, including J.D. Hayton (1960) who stated that davidite belonged to the group of multiple oxides which included arizonite and brannerite. He pointed out the difficulty in the analysis was because the oxidation state of the iron present was not known. Discussion continued, essentially confirming Hayton's opinion.

Today the internet offers numerous opportunities to find out about davidite, but care is needed to sort fact from fiction (or error), including misspelling of David's name, incorrect identification of Australian localities, and a considerable variety of compositions for davidite. However, the various internet sites regard it as a valid mineral, albeit with varying chemical analyses, while one site (`http://geo.oregonstate.edu/~taylore/minerals/davidite1938.htm`) shows a fine example of a crystal of davidite obtained from Radium Hill. Davidite, like David himself, has stood the test of time, and, despite some attempts to demote it, remains firmly fixed as an accepted mineral to this day.

EARLIER AUSTRALIAN WORK ON RADIOACTIVE MINERALS

There was some interest in the possibility of uranium being found in Australia as early as 1889 and even some vague reports of finds in the next couple of years (Mudd 2005). Mudd, based on Barrie (1982), mentions that G.W. Goyder (1826–1898) noted an unidentified green mineral at Rum Jungle as early as 1869, but it was not identified as a uranium mineral until 1917, when H.I. Jensen (1879–1966), a former student of David, reported uranium there.

What is probably the first specific mention of a radioactive mineral in Australia is the brief report, by G.W. Card (1865–1943) of the NSW Department of Mines, in 1894. He was referring to a small specimen of torbernite from a cobalt prospect at Carcoar, NSW (Card 1894). Edgeworth David had reported on the Carcoar prospect in 1888, when with the NSW Geological Survey. He commented on the variety of unusual minerals present, but did not find any uranium species (David 1888). Rayner (1957) in a fine review of the development of uranium work in New South Wales, refers to a report by A. Selwyn Brown (1898) on a radioactive mineral specimen said to come from Pambula on the far south coast of NSW. Rayner (1957) suggests that a more likely source of the sample was the Whipstick molybdenite deposit, some kilometres inland from the coast.

The next Australian radioactive mineral work occurred in April 1901 when Bernard F. Davis gave W.G. Woolnough (1876–1958), Demonstrator in Geology at Sydney University, and Edgeworth David a specimen he had collected in the Pilbara region of Western Australia. Woolnough identified it as gadolinite, a mineral containing a considerable percentage of rare earths. Davis took the specimen to England where he analysed it and sent back the results, which David and Woolnough presented to the Royal Society of New South Wales. The mineral was reported to have given off helium. Davis also collected two minerals 'allied to "euxenite" ... essentially niobates and titanates (with tantalum) of uranium, iron and yttrium earths with the cerium earths and thorium', (Davis 1902).

Of more significance, was a joint study carried out in 1904 by Mawson and T.H. Laby (1880–1946). Mawson had made his first contact with Edgeworth David in 1899 when in the first year of his Mining and Metallurgy Engineering Course at Sydney University. Even before his graduation on 19 April 1902, with Archibald Liversidge, Professor of Chemistry approving, Mawson was appointed a Junior Demonstrator in Chemistry, David acting as a referee (Branagan & Holland 1985, Ayres 1999). At the same time Mawson was studying to obtain his B.Sc. With Mawson was Acting-Demonstrator T.H. Laby who had been recommended by his chief, F.B. Guthrie (1861–1927) at the New South Wales Department of Agriculture's laboratory. Guthrie acted as Professor during several periods of leave by Liversidge (Branagan & Holland 1985). Laby was in the unfortunate situation of being unmatriculated, but he undertook night lectures in physics, chemistry and maths.

Mawson and Laby (1904), excited by the interest in radioactivity among chemists worldwide, set out to examine 'the more common [Australian] uranium and thorium minerals' first testing them photographically and then in an electroscope, based on the design of C.T.R. Wilson. Mawson built this instrument in the University's Engineering Laboratory. They tested some twenty Australian samples and, for comparison, three from overseas (Table 1). Most were monazite-rich sands, containing thorium, but two samples, one of torbernite from Carcoar, the other euxenite from Marble Bar, were uranium-bearing and highly active. The two researchers were particularly concerned to see if radium was given off by their samples, recognising it occurred in monazite from the Pilbara and another sample from Emmaville, NSW.

As neither of the authors was a member of the Royal Society of New South Wales, David made the formal presentation of the paper to an interested audience of the Society on 5 October, 1904 (Royal Society of NSW Proceedings, 1906), when Mawson knew he had his B.Sc. and was off shortly to Adelaide University as Lecturer. Meanwhile, Laby, despite his lack of a degree, had been awarded an 1851 Exhibition to study in England (Branagan & Holland 1985). Discussion followed the talk with David, Guthrie, James Pollock (1865–1922), Professor of Physics at the University, G.H. Knibbs (1858–1929), W.M. Hamlet (1850–1931) and the authors participating. The published version gave tribute to David for his encouragement, and also to Knibbs (at that time Lecturer in Surveying, later first Commonwealth Statistician), and J.A. Schofield (1869–1934), then Acting Professor in Chemistry.

Mineral	Activity	Locality etc.
Black uranium oxide, U_2O_5	100	Taken as standard
Torbernite	highly active	Carcoar, NSW (insufficient for comparative test)
Euxenite	highly active	Marble Bar tinfields, WA (insufficient for comparative test)
Gadolinite	0.88	Cooglegong River–Greenbushes tinfield, WA
Monazite	11.30	Pilbara, WA
Fine river sand with gold, tinstone, etc	8.49	Tumberumba, NSW.
Zircon sand with monazite	0.60	Tooloon River, NSW (contains 0.45% thoria)
Concentrated beach sand	7.39	Broken Head, Richmond River, NSW.
Concentrated river sand	8.00	Tasmania
Monazite	5.47	Torrington, NSW. A large well-developed crystal
Monazite	3.25	Torrington, NSW.
Monazite	4.92	Cow Flat, Torrington, NSW (contains 0.3% thoria)
Monazite	3.11	20 miles W of Torrington (contains 1.5% thoria)
Monazite	4.46	20 miles W of Torrington (contains 1.8% thoria)
Monazite	3.31	Gulf mine, Emmaville, NSW (contains 0.6% thoria)
Monazite	3.00	as above
Monazite	3.41	as above
Monazite	3.13	as above
Monazite	2.50	as above
Monazite	4.50	Paradise Creek, Emmaville
Pitchblende	354.05	Joachimstal (for comparison)
Samarskite	47.10	Sweden (for comparison)
Thallium blende	3.75	Locality unknown. Activity may be due to the presence of other uranium minerals

Table 1. Observations on radioactivity. Total activity as determined by ionisation produced in an air-gap. Modified slightly from the original table by Mawson & Laby (1904).

Mawson's work on radioactivity, from this early beginning, moved to the mineralogical, as noted above. In fact, it was not long after Mawson arrived in Adelaide that he paid a visit to the Moonta mines (Mawson, 1944) where S. Radcliff, a chemist, was testing the ore bodies for radium minerals (Radcliff 1906). Radcliff's work on the theoretical side was being assisted mainly by W.H. Bragg (1862–1942) at Adelaide University, who presented Radcliff's results to the local Royal Society, and Mawson's visit gets no mention by Radcliff. Bragg was heavily involved, at the time, in studying radium, uranium and thorium, presenting a series of papers the same year (Bragg 1906a, 1906b, 1906c), following on his Section A Presidential address on ionisation at the Australasian Association for the Advancement of Science (Bragg 1904). Radcliff (1913) continued his interest in radium, extracting the element from Radium Hill material. In this paper Radcliff suggests that Mawson took a parcel of 30 tons of picked ore from Radium Hill to the U.K. (sending some on to the USA), presumably when he provided a large sample for Crook and Blake, but elicited no interest among potential customers for the ore.

Laby continued in a chemical vein, working during his research at the Cavendish Laboratory, Cambridge under J.J. Thomson, for which he was awarded a B.A. (Close, 1983). In 1909 Laby, just as he was about to take up a position as Professor of Physics at Victoria University College, Wellington, New Zealand, offered another paper to the Royal Society of New South Wales. David was just back from the Antarctic, but the paper was read by Guthrie, Laby's first chief. The title is somewhat enigmatic: 'On a Pitchblende probably occurring in New South Wales' (Laby 1909). The paper must have been sitting around for a few years, as Laby had carried out the analysis while at Sydney University. It essentially continued on from the joint paper he had published with Mawson in 1904. The enigma is that the sample, passed from a mineral collector, Bennett in Newcastle, NSW, to Card, and thus to Laby, was thought to have come from the New England district of that State. However Laby cast doubt on this provenance when he mentioned in a footnote that a French metallurgist, C. Poulot, saw a large parcel of what he thought was the same ore at a treatment works in Germany. It had apparently been shipped from Melbourne, suggesting that New England was an unlikely source. As far as I am aware the true source was never established.

In the meantime, chemical research on radioactivity continued at Sydney University. George J. Gray, a student of David, graduated in Science (with Geology) in 1905; he also gained an Engineering Degree and went on to practise as a geologist (Branagan 1973, Vallance 1995). In 1907 Gray described work on the radioactivity of thorium carried out at the University on specimens once again obtained through Card. Gray (1907) thanked Liversidge for his encouragement. It was the year Liversidge retired and shortly after left for England, never to return.

E.S. Simpson (1875–1939) was another of David's Engineering students, graduating with honours in 1895, and working, over the years, largely on the minerals of Western Australia. Prider (1988) believed Simpson's 'best-known scientific contributions were in connexion with the rare radioactive minerals of the Pilbara' (Simpson, 1907, 1909, 1910, 1911, 1912a, 1912b).

ANOTHER SYDNEY UNIVERSITY ASPECT OF RADIOACTIVITY

In the years following the discovery of radioactivity, one of the major interests in uranium-bearing minerals was the perceived possibility of using the rate of breakdown to measure the age of rocks bearing significant quantities of radioactive minerals, and ultimately, in measuring the age of the Earth. David saw the possibilities on this work and, following World War One, urged Mawson to take up the problem testing Australian specimens. However he had too many irons in the fire until 1944 when he mentioned that he intended to use davidite from Radium Hill for an age determination, but he never seems to have got around to it.

In the meantime, through David's encouragement, the task was taken up in Australia by Leo Cotton (1883–1963), David's successor in the Geology Chair at Sydney University. Cotton, probably with the help of Harry Gooch (ca 1884–1946), the Department's jack-of-all-trades, built the equipment to carry out the uranium/lead method of dating rocks in the early 1920's. In particular, Cotton tested some Precambrian rocks from South Australia. During this period David and Cotton were in close touch with Arthur Holmes (1890–1965) in England (David Papers, University of Sydney Archives), who had established his reputation as an authority on such matters. They were particularly interested in the methods of calculating rock ages, and Cotton did not accept Rutherford's Constant used by Holmes, arguing for a slight revision. In the end Cotton's dates differed only slightly from those of Holmes for samples from the same rock body (Cotton, 1926). Cotton first presented his results at a lecture in Adelaide, before sending off his paper to the *American Journal of Science* (Table 2). He followed up this work several years later (Cotton 1928), discussing measurements of the age of the Earth.

David mentioned the radioactive minerals in his 1932 *Explanatory Notes* (David 1932), but surprisingly or not, davidite does not get a mention, the nearest statement being 'radio-active ilmenite'. Did David suspect it wasn't a 'fair dinkum' mineral? The additional notes about these minerals in the *Geology of the Commonwealth*, which finally appeared in 1950, probably came not from David's pen, but from his former pupil, as editor, W.R. Browne (David 1950). Completed in the years immediately after the War Browne (Vol. 2, p. 315) gives davidite at Radium Hill only a minor place among the ore minerals. As mentioned above, just a few years later, when mining began, it was found to be the major uranium mineral in the ore body.

Mineral	U_3O_8	UO_3	UO_2	ThO_2	PbO	Age (Ma)[a]
Fegusonite, Coolglegong, WA		2.38		0.53	0.18	620
Mackintoshite, Wodgina, WA		Present		24.72	7.90	1475
Thorogummite, Wodgina, WA		37.33	35.6	24.46	7.78	1460
Pilbarite, Wodgina, WA		27.09	none	31.34	17.26	3840
Concentrates, Olary, SA	1.6		none		0.16	880
Carnotite, Olary, SA					1.3	240
Lodestuff[b], Olary, SA	47.8				0.40	1560
Monazite[c], Normanville, SA	2.25			10.70	0.55	1130

Table 2. The lead, uranium and thorium content (percentages) of certain Australian radioactive minerals. Modified slightly from the original table by Cotton (1926). Notes: (a) Pb/U+0.384 Th × 8000. (b) An 'intimate mixture of ilmenite, rutile and magnetite with a variable but small amount of other material' (Crook & Blake 1910). (c) R.G. Thomas, using a slightly different formula, obtained a value of 1073 Ma. For references to the original samples see Cotton (1926).

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David Branagan
School of Geosciences, University of Sydney
dbranaga@mail.usyd.edu.au

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